Particle Emissions from Compressed Natural Gas Engines

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This paper presents the results of measurements conducted to determine particle and gas emissions from two large compressed natural gas (CNG) spark ignition (SI) engines. Particle size distributions in the range from 0.01-30 μm, and gas composition were measured for five power settings of the engines: 35%, 50%, 65%, 80% and 100% of full power. Particle emissions in the size range between 0.5 and 30 μm, measured by the Aerodynamic Particle Sizer (APS), were very low at a level below 2 particles cm^{-3}. These concentrations were comparable with average ambient concentration, and were not considered in the succeeding analysis. Both engines produce significant amounts of particles in the size range between 0.015-0.7 μm, measured by the Scanning Mobility Particle Sizer (SMPS). Maximum number concentrations of about 1×10^{7} particles cm^{-3} were very similar for both engines. The CMDs were in the range between 0.020 and 0.060 μm. The observed levels of particulate emission are in terms of number of the same order as emissions from heavy duty diesel engines (Morawska et al. 1998). On the other hand emissions of CO and NO_x of 5.53 and 3.33 g/kW-hr, respectively, for one of the tested engines, were considerably lower than set by the standards. According to the specifications for the gas emissions, provided by the US EPA (US EPA, 1997), this engine can be considered as a "low emission" engine, although emissions of submicrometer particles are of the same order as heavy duty diesel vehicles.

1. Introduction

In recent years fine and ultra fine particles emitted from internal combustion engines have attracted an increasing level of attention. This attention has arisen from epidemiological studies conducted by a number of research groups and pointing to the health effects resulting from inhalation of fine particles (Dockery et al. 1993; Pope et al. 1992; Schwartz & Dockery

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1992; Wichmann et al. 1989; Schwartz & Marcus 1990; Ostro 1984). Among other investigations, recent studies on particles smaller then 0.1 μm, indicated that they can cause health effects by virtue of their size. In particular, in some cases they can cause stress of the epithelial cells lining the lung, leading to irritation and inflammation (Ferin & Penney 1992; Donaldson et al. 1996). As a result of the findings and indications from these studies, health and environmental policy bodies have directed an increasing level of interest to this area (Chow et al. 1994; Department of the Environment 1994).

The emissions from internal combustion engines have been regulated solely on the basis of total particulate mass emission; there is no reference to the size of the particles or to the number concentration of emitted particles. To comply with the current standards, vehicle and engine manufacturers have been improving engine designs, which has resulted in lowering of total particulate mass emissions. The emissions of other regulated pollutants such as NO_x and CO were also significantly reduced. Unfortunately, these reductions may have been accompanied by a dramatic increase in the particle number emissions, especially in the very fine particle size range, below 0.1 μm (Johnson & Baumgard 1996).

Over the past twenty years effort has been devoted to the measurement and characterisation of particle emissions from diesel engines. There have not been many investigations focused on spark ignition engines, and consequently there is significantly less information available on particles from spark ignition (SI) emissions. It is only recently that there has been some information published about the effects of vehicle and fuel type on particle size from spark ignition engines (Ristovski et al. 1998; Rickeard et al. 1996; Greenwood et al. 1996; Graskow et al. 1998). There is evidence from recent studies, that the number weighted emissions of SI engines at relatively high loads are comparable to those from diesel engines (Greenwood et al. 1996; Rickeard et al. 1996). High emission levels from SI engines were also observed during the so called “spikes” in the exhaust particle concentrations (Graskow et al. 1998). The observed exhaust particle concentrations during these spikes increased by as much as two orders of magnitude over the baseline concentration and were up to 10^7 particles/cm ^{3}. The particle “spikes” are formed through the homogeneous nucleation of gas phase heavy hydrocarbons. The source of these hydrocarbons is uncertain, though it is believed that they are associated with the occasional break-up of intake valve and combustion chamber deposits. Even less data is available on particle emissions from CNG fuelled SI

engines. An isolated study in this area by Greenwood et al. (1996), indicated that the particle concentration levels emitted from CNG fuelled engines are similar to gasoline engines, and at high loads approach those of diesel engines.

The aim of the work presented here was to investigate size distribution and concentration levels of particles emitted from two large four stroke spark ignition gas engines, as well as composition of gas emissions. The results obtained are of importance for understanding of the nature of emissions and for estimating the contribution of gas engine emissions to the overall particle pollution levels. The obtained results were also interpreted with a view to provide a qualitative explanation as to the mechanisms of particle transformation processes immediately after generation by the engines.

2. Experimental Methods

2.1 Engines

Two large four stroke spark ignition gas engines located at Southside Engineering, Brisbane, Australia, were used for the testing. The characteristics of the engines are presented in Table 1. The first engine was of an older type (Waukesha 5115GL Constant Torque Engine) without any emission control, while the second (Deutz, model TBG 616 K) was a modern computer controlled engine with emission controls.

The basic aspects of the computer-controlled emission can be summarised as follows. Increase in the proportion of excess air can impede the formation of nitrogen oxides produced by reactions at high temperatures. With too much excess air, the combustion temperature drops, which results in an increase in concentrations of emitted CO and incompletely burnt hydrocarbons (C_mH_n). Furthermore, the efficiency decreases as a result of decreasing combustion velocity. The system for electronic mixture control of the Deutz engine varies the excess air factor in order to keep the emission of both NO_x and CO as well as C_mH_n low, and at the same time to obtain optimal efficiency. For this purpose, the mixture control system uses, among other parameters, the mean process or combustion chamber temperatures of all cylinders. For natural gas operation, an oxidation catalytic converter is generally used. The engine adjusts itself automatically for each load condition.

The older type Waukesha engine did not have automatic adjustment, and had to be adjusted manually for each load condition. This was done by monitoring the excess oxygen and CO_2 concentrations in the exhaust.

2.2 Fuel

Both engines used the same type of natural gas supplied by BOC. Table 2 lists the concentrations of various constituents in the gas, as obtained by the supplier.

2.3 Dilution and Sampling System

The main factors considered in the design of the sampling port were: high temperature of the exhaust, potentially very high concentration of particles in the exhaust, potential non-uniformity of exhaust parameters as a function of the geometrical cross section through the pipe, and isokinetic sampling conditions. As most of the particle emissions were expected to be in the range below 4 μm, it was not expected that lack of isokineticity of sampling would influence the results (Kittelson et al. 1980). Nevertheless care was taken to achieve isokinetic sampling conditions.

The exhaust samples for particle measurements were drawn from the exhaust manifold through a 3.8 mm I.D. stainless steel probe which faced into the exhaust stream in the centre of the exhaust pipe. The flow rates at which samples were collected from the exhaust varied between 24 to 61 lpm, for 35 and 100% load respectively. Sampling from the centre of the exhaust pipe ensured that the sampling geometry was well defined and could be reproduced in the future. The sample was then directed to a dilution tunnel where it was mixed with clean air at ambient temperature of 25°C and 47%RH. Mixing of the exhaust sample with clean air in the dilution tunnel served two purposes. Firstly, the diluted exhaust was cooled to temperatures at which it could be sampled for further analysis, and secondly, particle concentrations were decreased to levels suitable for particle sizing instrumentation. Particle concentration decrease in the dilution process taking place in the dilution tunnel is effective in decreasing particle coagulation rate. This is believed to be representative of real world engine operation as after emission, particles rapidly mix with ambient air (Morawska et al. 1998). The exhaust sample should not be cooled down before it is diluted, as rapid cooling of a concentrated aerosol may result in significant changes of its physico-chemical characteristics (Kittelson et al. 1980).

All particle sizing instruments sampled directly from the dilution tunnel. The flow of clean air was adjusted to 160 L min^{-1} in order to achieve isokinetic sampling conditions from the dilution tunnel. Details of the dilution tunnel were published elsewhere (Morawska et al. 1998).

2.4 Particle Sizing Instrumentation

Two different instruments were used for particle size measurements. Particle size distributions were measured by the TSI Model 3934 Scanning Mobility Particle Sizer (SMPS) in the size range 0.015-0.7 μm, and by the TSI Model 3030 Aerodynamic Particle Sizer (APS) in the size range 0.5-30 μm. The operating principles of these instruments are described elsewhere (Knutson & Whitby 1975; Wang & Flagan 1990; Remiarz et al. 1983).

Calibration of the SMPS and APS instruments is performed routinely by the measurement of aerosols of known sized distributions, such as PSL spheres. The SMPS instrument is also routinely cross-validated against a second SMPS located at the QUT Environmental Aerosol Laboratory.

2.5 Exhaust Gas Composition

Composition of the exhaust gases, elemental composition of particles and Polycyclic Aromatic Hydrocarbons (PAH) measurements were performed only for the Deutz engine. Two instruments were used for exhaust gas composition analysis: a Stack Gas Analyser (IMR 3000P) and combustion efficiency analyser (Telydine Max5). The Stack Gas Analyser was used for analysis of the following gases: excess oxygen (O_2) up to 30%; carbon dioxide (CO_2) up to 20%; carbon monoxide (CO) up to 2000 ppm; sulphur dioxide (SO_2) up to 2000 ppm; and nitric oxides (NO_x) up to 1000 ppm. The combustion efficiency analyser measured incompletely burned hydrocarbons up to 5% with a resolution of 0.2%.

2.6 Experimental Methodology

As no standards for emission measurements of this type of engine are available, measurements of size distribution and concentration of particles were performed for five power settings of the engine: 35%, 50%, 65%, 80% and 100% of full power. Three consecutive samples were taken at each setting with the SMPS and, five with the APS. Time needed for one sample with the SMPS was 90 s and with the APS 20 s. The engines were cycled up from the lowest power setting to the highest and then down again to allow repeat

sampling for the same power setting. There were no measurements conducted of the main exhaust flow rates, and the values provided by the manufacturer of the gas engine for each power setting, were later used for data analysis. The flow rates supplied by the manufacturer, in units of mass per unit of time (kg h^{-1}), were normalised to atmospheric pressure and temperature to unit of volume per unit of time (m^3s^{-1}).

Exhaust gas composition measurements were performed at the same time as particle size measurements. The data was continuously logged on a computer and averaged values, together with standard deviations, were calculated for each mode and each analysed gas.

3. Experimental Results and Discussion

3.1 Particle Number Size Distribution Measurements

Particle emissions in the APS range (0.5-30 μm) were very low, at a level below 2 particles cm^{-3} which is comparable with an average ambient concentration, and hence were not considered in the following analysis.

Analysis of particle engine emissions should include both the nature of the particles and the total number emitted. Most of the observed size distributions were unimodal and lognormal, and thus CMD was chosen to represent particle size and trends in size distribution. Particle number size distributions for Waukesha and Deutz engines, for different power settings, are presented in Figure 1 and Figure 2 respectively, while the summary of the measurements in terms of particle concentration, count median diameter (CMD) and geometric standard deviation is provided in Table 3. Arrows in Figure 1 and Figure 2 indicate the time order in which the measurements were conducted. A significantly smaller number concentration of particle emissions were measured at 50% and 35% of power, down cycle for the Deutz engine compared to the Waukesha engine. This was due to the decrease in particle diameter below the measuring range of the SMPS, as a result of which a certain number of particles were not measured. The instrument registered the upper part of the particle distribution, while the majority of the emitted particles were below 0.015 μm. Thus the measurements at 50% and 35% power were not considered in further analysis and are not included in Figure 2. Figure 3 presents the dependence of the particle CMD on engine power for both engines, while Figure 4, presents the total particle number concentration corrected for the dilution

factor. Arrows on both figures indicate the time order in which the measurements were conducted.

As can be seen from Figures 3 and 4, the two analysed CNG engines show clear trends both in particle concentration and in particle size distribution, characterised by their total number concentration and CMD, respectively. For both engines maximum number emissions were achieved for 100% power. It is interesting to note that both engines had very similar maximum number concentrations of about 1×10^{7} cm^{-3}. While the Deutz engine had minimum emission for minimum power, the Waukesha engine had minimum emission for 50% power for both cycles (up and down). There was, approximately, a factor of six difference between minimum and maximum in particle emission concentrations which occurred at 50% and 100%, for the Waukesha engine, and 35% and 100% of maximum power, for the Deutz engine.

The CMDs were in the range between 0.020 and 0.060 μm for both engines and for all 5 modes, except for the second engine at 65% of power. This would indicate that all peaks belonged to the nucleation mode, with the exception of the peak at 0.140 μm generated by the second engine at 65% power, which belonged to the accumulation mode. The shift displayed by the second engine could be due to excess fuel intake that led to the formation of aggregates. Bimodal size distributions, not included in this analysis, were observed only during the process of manual tuning of the first engine. The hypothesis is that during the tuning process fuel was in overabundance and led to the formation of two peaks, one in the nucleation mode and one in the accumulation mode.

The number size distribution exhibits an approximately linear dependence on engine load for loads greater than 50% for the Waukesha engine and in the whole range for the Deutz engine. When the engine load increases, the number of particles in the smaller size range decreases, while the number of particles in the higher size range increases. The total number concentration also increases by almost an order of magnitude. If the particle concentrations achieved at higher loads were sufficiently high for coagulation growth to occur, especially of small particulates, a reduction in number, accompanied by an increase in particle size would have been observed. As there was an increase in both the size and the number concentrations measured, a coagulation model can not describe the process taking place. It is more likely

that a situation where coagulation together with homogenous nucleation of metallic ash particles, and heterogenous nucleation of PAH and carbon soot on the metallic ash particles took place at the same time. These hypotheses would require further investigations to be validated.

There is no literature data available on the size distribution of particles for similar types of engines. The only available data are for light duty vehicles run on compressed natural gas (Greenwood et al. 1996). The size distributions measured here are in general, similar to those reported by Greenwood et al. 1996, although the CMD's reported in this work are smaller. The other main difference is that in the previous measurements (Greenwood et al. 1996) most of the particles were larger than 0.020 μm, while in the measurements reported here, for small load conditions, a significant number of particles smaller than 0.020 μm was observed.

Previous studies on spark ignition engines (Graskow et al. 1998) observed particle size distributions that were very poorly represented by a lognormal distribution function. The authors concluded that the formation mechanisms of aerosols from spark ignition engines might differ from those of diesel engines, which are well represented by lognormal distributions. To test this hypothesis a typical particle size distribution from the measurements reported here was fitted with a lognormal distribution function. Figure 5 presents a typical number size distribution measured by the SMPS at 100% load conditions for the first engine, as well as a very close lognormal fit of the distribution, with a peak at about 0.060 μm. Similarly close fits by lognormal distribution functions were observed for other spark ignition engines, and also for other types of fuel (LPG, unleaded and leaded petrol) and were published elsewhere (Ristovski et al. 1998). This suggests that the formation mechanism of most spark ignition aerosols could be similar to the diesel engine aerosols. The discrepancy between the lognormal distribution function and previously measured size distributions (Graskow et al. 1998) can not be generalised to all spark ignition engines, and could be a characteristic of the particular engine that was tested.

Particle number emission data have been reported in the literature for a number of different Diesel and SI engines (Graskow et al. 1998; Ristovski et al. 1998; Morawska et al. 1998). In order to compare these results to those in the literature, number emissions of both investigated gas engines were expressed in terms of the brake specific particle number

(BSP_N), which has units of number of particles emitted per engine kilowatt hour. Table 4 presents comparison between particle emissions from the engines tested here versus those reported in the literature (Morawska et al. 1998).

Table 4 reveals that CNG engines at all tested loads and diesel engines have similar levels of submicron particle emissions. SI engines running on unleaded petrol achieve these high levels only on highest modes (120 km/h) (Graskow et al. 1998). However, the power of the investigated gas engines are ten and five times higher, respectively, than the power of a diesel bus, and these engines are used on full or almost full load most of the time. Thus these gas engine's submicron particulate output is, roughly, the equivalent of five to ten buses running on full load. The other difference between diesel combustion emissions and gas combustion emissions is in the particle size distribution, and changes to the particle distribution with the change of power. While for diesel emissions there is a significant number of particles in the upper submicron range, with particles larger than 1 μm emitted at high power settings and high loads (Morawska et al. 1998), all the emissions for gas combustion are in the lower submicron range.

3.2 Measurements of Gas Composition

Table 5 presents experimental and calculated data on the relationship between engine power settings and gas composition of the exhaust. The concentrations of hydrocarbons were below the detection limit (< 100 ppm) of the instrument and are not presented in Table 5. In the last column a sum of NO and NO_2 is presented as nitrogen oxides (NO_x). Values for CO_2 and O_2 are in percentages while for all other gases are in parts per million (10 000 ppm = 1%). The average values and standard deviations, over the whole testing cycle, for each gas are also presented.

It can be seen from the data presented in Table 5, that emissions of CO_2 and O_2 were the most consistent of all the gas emissions. The difference in the concentration of CO_2 or O_2 for modes with the same power load was smaller than 0.1%. Other gases, such as CO and NO_x, did not show any trend and their values fluctuate around the mean value of 412 ppm for CO and 214 ppm for NO_x. The standard deviations for all nine measurements are 2.2% and 5% for CO and NO_x, respectively. Sulphur dioxide (SO_2) shows a decreasing trend from 84 ppm for 35% power up, to 22 ppm for 80% power down, (measurement no.6) where it remains

constant through to the last mode (measurement no.9) at 35% power load. As yet there is no available explanation for the trend in the emission of SO_2. The average values of the emissions in terms of mass per unit volume were 0.515±0.011 g m^{-3} for CO and 0.310±0.020 g m^{-3} NO_x. These values are smaller than the values given by the manufacturer, which are: 0.650 g m^{-3} for CO and 0.5 g m^{-3} for NO_x.

4 Conclusion

Although there are no standards available for non-road spark ignition engines greater than 25 HP, it would be illustrative to compare the emission of the tested engine with the existing standards for heavy-duty highway spark ignition engines (US EPA September 1997). Standards are given only for the so-called regulated pollutants: CO, HC, and NO_x, and are expressed in grams per kilowatt-hour. Table 6 provides a comparison between the gas emission levels of the second engine and the levels set by the standards. It can be seen from Table 6 that emission of CO and NO_x are considerably lower than set by the standards and according to the specifications provided by the US EPA (US EPA September 1997) this engine can be considered as a “low emission” engine.

Although the manufacturers have significantly reduced the emissions of gases, number emissions of nanoparticles (particles smaller than 0.1 μm) for these engines are very high and of the same order as diesel engines. While for diesel engines standards on particle emission exist at least for the total emitted mass, there are no standards for SI engines. However, if nanoparticle emissions will prove to constitute a significant health risk emissions from CNG engines will need to be regulated, as they present a significant source of these particles.

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Figure Captions

Figure 1. Particle size distributions for different load conditions, engine 1.

Figure 2. Particle size distributions for different load conditions, engine 2. Size distributions for the last to modes (50% and 35% down) were excluded.

Figure 3. Dependence of the count median diameter on the engine load.

Figure 4. Relationship between the total number concentration in the exhaust and the engine power. The arrows indicate the time order in which the measurements were conducted.

Figure 5. Typical size distributions measured by the SMPS at 100% load conditions for the first engine. A fit to a lognormal distribution is presented with a full line. Parameters used to reproduce the solid line are: geometric mean diameter $d_g = 59\text{nm}$, and geometric standard deviation $\sigma_g = 1.46$.

Table 1 Characteristics of the tested engines

	Engine 1	Engine 2
Model	Waukesha 5115 GL	Deutz TBG 616
Cylinders	V-12	V-12
Piston Displacement [litters]	84	26.3
Power Rating [kW]	1082	525
Speed Range [1/min]	1500	1500

Table 2. Composition of the Compressed Natural Gas

Component	Specification [%]
Carbon Dioxide (CO_2)	0.58
Oxygen (O_2)	0.0
Nitrogen (N_2)	4.7
Methane (CH_4)	88.21
Ethane (C_2H_6)	5.59
Propane (C_3H_8)	0.59
I-Butane (C_4H_{10})	0.19
N-Butane (C_4H_{10})	0.07
I-Pentane (C_5H_{12})	0.04
N-Pentane (C_5H_{12})	0.02
C_6s’	0.01
Total	100

Table 3 Relation between particle size and number distribution and the engine power.

	Engine 1		Engine 2	
	Up ↑	Down ↓	Up ↑	Down ↓
At 35 % Power [kW]	356		184	
CMD [nm]	60.4	72	25	<17
Standard Deviation	1.4	1.4	1.39	
Total No. [cm^{-3}]	4.53E6	5.138E6	2.49E6	2.02E4
At 50 % Power [kW]	534		263	
CMD [nm]	36.03	47	26.3	<17
Standard Deviation	1.42	1.41	1.6	
Total No. [cm^{-3}]	1.5E6	3.529E6	8.42E6	3.63E4
At 65 % Power [kW]	711		341	
CMD [nm]	36.69	49	137.7	35
Standard Deviation	0.73	1.65	1.72	1.39
Total No. [cm^{-3}]	3.78E6	2.293E6	9.26E6	4.27E6
At 80 % Power [kW]	889		420	
CMD [nm]	46.35	55	51.08	39.6
Standard Deviation	3.62	1.55	1.5	1.41
Total No. [cm^{-3}]	7.81E6	6.797E6	1.07E7	7.01e6
At 100% Power [kW]	1067		525	
CMD [nm]	58.12		57.8	
Standard Deviation	1.52		1.46	
Total No. [cm^{-3}]	1.07E7		1.2E7	

Table 4 Comparison between particle emissions from the CNG engines tested versus diesel engines reported in the literature (Morawska et al. 1998).

Power settings for gas engines [%]	BSP_N Engine 2 [part kW^{-1} h^{-1}]	BSP_N Engine 1 [part kW^{-1} h^{-1}]	BSP_N Diesel [part kW^{-1} h^{-1}]	Power settings for diesel engines [%]
35	1.20E+13	1.08E+14	2.42E+14	10
50	4.75E+13	3.41E+13	1.39E+14	25
65	7.42E+13	5.92E+13	9.58E+13	50
80	9.65E+13	9.34E+13	1.02E+14	75
100	1.30E+14	1.43E+14	1.26E+14	100

Table 5 Relationship between engine power and the gas composition in the exhaust for the second engine.

Power Setting (%max)	Meas. No.	Carbon Dioxide (% CO2)	Excess Oxygen (% O_2)	Carbon Monoxide (ppm CO)	Sulfur Dioxide (ppm SO_2)	Nitric Oxide (ppm NO)	Nitric Dioxide (ppm NO_2)	Nitric Oxides (ppm NO_x)
30.7	1	7.26	8.13	428	84	195	5	200
50	2	6.99	8.54	416	61	181	25	206
65	3	6.82	8.76	412	46	188	33	221
83	4	6.75	8.85	409	24	198	34	232
100	5	6.60	9.20	418	31	162	37	199
80	6	6.79	8.89	401	22	179	43	223
65	7	6.83	8.73	400	22	175	42	217
47.6	8	6.93	8.57	418	23	170	44	214
35	9	7.10	8.31	406	23	162	47	209
	Av.	6.86	8.73	412	41	183	31	214
	SD	0.20	0.32	9.18	22.24	13.02	13.02	10.75

Table 6 US-EPA Standards for Heavy-Duty Highway Spark Ignition Engines

	CO [g/kW-hr]	NO_x [g/kW-hr]
Engine 2	5.53	3.33
US EPA standard (≤14,000 lbs.)	19.3	6.7
US EPA standard (>14,000 lbs.)	50.	6.7